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Identification of the $J^\pi = 1^-$ state in ^{218}Ra populated via α decay of ^{222}Th

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The α decay of ^{222}Th populating the low-lying $J^\pi = 3^-$ state, and also a proposed 1^- state, in ^{218}Ra has been observed. The observations suggest an excitation energy of 853 keV for the 1^- state, which is 60 keV above the 3^- state. The hindrance factors of these α decays give a possible boundary to the region of ground-state octupole deformation in the light-actinide nuclei. The relative positions of the $J^\pi = 1^-$ and 3^- states suggest they are produced by an octupole-vibrational mechanism, as opposed to α clustering or rotations of a reflection-asymmetric octupole-deformed shape.

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I. INTRODUCTION

The phenomenon of octupole collectivity in atomic nuclei has been a topic of theoretical and experimental investigation for over half a century [1–6]. Proton and neutron orbitals with $\Delta l = \Delta j = 3$ give rise to an enhancement of the octupole part of the nucleon-nucleon interaction. This can result in collective behaviour such as octupole vibrations or, in nuclei with stronger octupole correlations, reflection-asymmetric octupole deformations, with possible evidence for the latter case recently obtained from direct measurements of $B(E3)$ strengths [7]. The part of the nuclear chart where the largest octupole correlations are expected is the light-actinide region around $N \sim 134$ and $Z \sim 88$. The ground states of nuclei in this region have been predicted to evolve from spherical at $N = 130$, to quadrupole-octupole deformed around $N = 134$ before possessing just quadrupole-deformed shapes close to $N = 140$ [8]. Experimental determination of the boundaries of this possible octupole-deformed region is important to guide theoretical predictions of the strength of octupole correlations and validate those which agree.

The $N = 130$ nucleus ^{218}Ra is of interest in this regard since it lies in the transitional region between the spherical nuclei, just above the $N = 126$ neutron shell closure, and the region of nuclei with expected octupole components of their deformation at $N \sim 134$ and $Z \sim 88$. The ground state of ^{218}Ra is expected to be spherical [8], but it has been shown that at high spins, the yrast states form an alternating-parity sequence, with enhanced $E1$ transitions between the positive- and negative-parity states [9–13]. This high-spin structure is characteristic of strong octupole correlations, and it has been suggested that the octupole shape in this nucleus is stabilised by rotation [12, 14]. The low-lying negative-parity states in ^{218}Ra, specifically, those with $J^\pi = 1^-$ and 3^-, can thus offer valuable insight into the development of octupole collectivity in this region as a function of N, and also as a function of angular momentum.

In the 1980s, Gai *et al.* [10] tentatively assigned the $J^\pi = 1^-$ state with an excitation energy of 713 keV, approximately 80 keV below the established $J^\pi = 3^-$ level. This observation was consistent with the interpretation that the states result from α-particle clustering, fitting well with the theoretical predictions of α-cluster models [15]. However, subsequent in-beam γ-ray spectroscopy experiments could not reproduce this observation; Wieland *et al.* [13] stated that no energy levels were present between the $J^\pi = 2^+$ and 3^- states, in contradiction to the level scheme proposed by Gai *et al.*, and that the $J^\pi = 1^-$ state, therefore, presumably lies above the $J^\pi = 3^-$ state. This ordering of the low-energy negative-parity levels would contradict the theory of α-particle clustering but would be consistent with an octupole-vibrational picture. Gai, however, replied [16] suggesting that the experiment performed by Wieland was not optimised to search for the $J^\pi = 1^-$ state and that their non observation was not enough to warrant their conclusions.

The new results presented in this paper are from the investigation of low-energy negative-parity states of ^{218}Ra, populated following the α decay of ^{222}Th. Previous studies of the α decay of ^{222}Th [17–21] have shown that it proceeds via a ground state (^{222}Th) to ground state (^{218}Ra) transition with $E_\alpha = 7980(2)$ keV [21] and via a ground state (^{222}Th) to $J^\pi = 2^+$ excited state (^{218}Ra) transition with $E_\alpha = 7599(2)$ keV [21]. In the present work, α decays from the ground state of ^{222}Th to the $J^\pi = 3^-$ and tentatively proposed 1^- states of ^{218}Ra have been observed for the first time.

II. EXPERIMENTAL DETAILS

In the present work ^{222}Th nuclei were produced in the fusion-evaporation reaction ^{208}Pb(^{18}O,4n)^{222}Th with a beam energy of 95 MeV, a target thickness of 0.45 mg cm^{-2} and a 0.1 mg cm^{-2} carbon degrader foil. The experiment was performed at the Accelerator Laboratory of the University of Jyväskylä, Finland. An average beam intensity of ∼18 pnA was used for a duration of ∼157 hours. The target was located at the centre of the SAGE spectrometer [22], which is used to detect prompt γ rays and internal conversion electrons; however, data from the SAGE spectrometer were not used for the results discussed here. The recoiling nuclei were separated from fission fragments and unreacted beam ions using the RITU gas-filled recoil separator [23, 24] and were subsequently implanted into two double-sided silicon-strip detectors (DSSDs), which are part of the GREAT spectrometer [25] located at a focal plane of RITU. The two DSSDs each consisted of 40 horizontal and 60 vertical strips giving a total of 4800 individual detector pixels. An array of 28 silicon PIN diode detectors were located upstream of the DSSDs positioned to detect charged particles that were emitted out of the DSSDs. A multi-wire proportional counter (MWPC), which is normally placed at the entrance of GREAT to measure the energy loss and time-of-flight of recoils, was not used in this work due to the low recoil energies of the ^{18}O + ^{208}Pb reaction products. An array of three HPGe clover detectors surrounding the DSSDs was used to detect γ and X rays emitted by decaying implanted recoil nuclei.

III. DATA ANALYSIS

The DSSDs were calibrated using α particles emitted by implanted nuclei, or those in their subsequent decay chains, produced during the experiment. The α particles used were from ^{210}Po [$E_\alpha = 5304.33(7)$ keV], ^{220}Ra [$E_\alpha = 7453(7)$ keV], ^{219}Ra [$E_\alpha = 7678(3)$ keV], ^{213}Rn [$E_\alpha = 8088(8)$ keV] and ^{221}Th [$E_\alpha = 8470(5)$ keV]. The absolute efficiency for the detection of γ rays in the focal-plane clover detectors as a function of γ-ray energy was established by comparing the intensities of α particles in the DSSDs with intensities of γ-ray transitions in $\alpha\gamma$-coincidence data.

The α decays of ^{222}Th nuclei were selected by correlating either two (recoil-α) or three (recoil-α-α) signals within a single pixel of the DSSDs. Chronologically, the signals corresponded to: (i) the recoiling ^{222}Th nucleus entering the DSSD; (ii) the α particle emitted following the decay of ^{222}Th, with a time gate up to 16 ms ($\sim$ seven half-lives); and (iii) the α particle emitted following the decay of ^{218}Ra, with a time gate set up to 180 μs ($\sim$ seven half-lives). When measuring the ^{222}Th α-particle energies, signals in the PIN detectors of GREAT were used to veto any coincident DSSD signals, hence removing from the spectra some of the partially deposited energies from escaping α particles. However, when using the ^{218}Ra α decays to identify a recoil-α-α chain, no PIN detector veto or energy gate was used so as to include the escaping α particles.

Signals in the DSSDs were assumed to be due to implanting recoils, and therefore vetoed as α decays, if a γ ray or conversion electron was detected in the SAGE spectrometer at a time preceding the DSSD signal. A two-dimensional gate was set for the veto over the recoils time-of-flight through RITU ($\sim$ 2 μs) and their energy distribution in the DSSD ($\sim$ 2 MeV). This somewhat compensated for the absence of a MWPC at the entrance of GREAT.

The data analysis was performed using the GRAIN software [26], which was developed for use with data acquired by the Total Data Readout system.

IV. RESULTS

Coincidences between α particles and γ rays were studied following the selection of a recoil-α chain and are shown in Fig. 1(a). The α particles of ^{222}Th were identified with the help of the diagonal lines shown on the $\alpha\gamma$-coincidence spectrum. The lines represent a constant energy for the sum of the α-decay Q value, calculated from the α-particle energy, and the γ-ray energy. They are equal to the Q values between the ^{222}Th ground state and the ground state, $Q[0^+(^{222}\mathrm{Th})\to 0^+(^{218}\mathrm{Ra})]$ (dashed line), and $J^\pi = 2^+$ state, $Q[0^+(^{222}\mathrm{Th})\to 2^+(^{218}\mathrm{Ra})]$ (dot-dashed line), of ^{218}Ra. In Fig. 1(a) the $\alpha\gamma$ coincidences assigned to three α decays of ^{222}Th to excited states in ^{218}Ra are circled and labelled, with contaminant coincidences from ^{213}Rn, ^{219}Ra and ^{221}Th also indicated. The α-particle energies, E_α, branching ratios, b_α, and hindrance factors, f, (defined in Section V A) of the four α decays identified from ^{222}Th along with the spins, parities and energies of states populated in ^{218}Ra are given in Table 1. Figure 2 shows the level scheme of ^{218}Ra populated by the α decay of ^{222}Th with the proposed $J^\pi = 1^-$ state included. In the present work, the half-life of the ^{222}Th ground state has been measured to be $T_{1/2} = 1.964(2)$ ms. This value is lower than the previous measurements of 4(1) ms [17], 2.8(3) [18], 2.2(2) [19], 2.0(1) [20] and 2.237(13) [21].

A. α decay to the ground state and $J^\pi = 2^+$ state in ^{218}Ra

The α decays from the ground state of ^{222}Th to the ground state and the 2^+ state at 389 keV of ^{218}Ra have previously been established, with α-particle energies of 7980(2) and 7599(2) keV, respectively [21]. In the present work, the α decay which directly populates the ground state of ^{218}Ra was observed with energy 7986(3) keV and branching ratio 98.16(5)%. Only random coincidences between these α particles and background γ rays were observed. The α decay to the 2^+ state of ^{218}Ra has been observed in the present work with energy 7603(3) keV and branching ratio 1.81(1)%. These 7603-keV α particles can be seen in Fig. 1(a) in coincidence with the 389-keV $2^+ \to 0^+$ γ ray. As expected, these coincidences appear on the $Q[0^+(^{222}\mathrm{Th})\to 0^+(^{218}\mathrm{Ra})]$ line.

B. α decay to the $J^\pi = 3^-$ state in ^{218}Ra

In Fig. 1(a) coincidences between ^{222}Th α particles with $E_\alpha = 7205(4)$ keV, and γ rays with energies 389 and 404 keV are indicated. The 7205-keV α particles are identified as being from ^{222}Th by their half-life and that of the subsequent ^{218}Ra decays. Figure 1(b) shows the γ rays in coincidence with the 7205-keV α-particles, contaminant coincidences are present from the escaping α decays of ^{219}Ra (316 keV) and ^{221}Th (331 keV). The intensity of the 389-keV γ ray in coincidence with the 7205-keV α particle is larger than that of the 404-keV γ-ray coincidences, when taking into account detector efficiencies and conversion coefficients. This is presumed to be due to false coincidences between 389-keV γ rays and the more abundant 7603-keV α particles which have escaped from the DSSD without depositing their full energy. From the spectrum of α-particle energies in coincidence with the 389-keV γ rays it is difficult to establish that 7205-keV α particles are present. However, $\gamma\gamma$ coincidence analysis of the spectrum in Fig. 1(b) reveals one coincidence event between the 389- and 404-keV γ rays in a virtually background free spectrum. This single count would be expected when considering the intensity of the 404-keV γ ray and the efficiency of the detector array.

As the $\alpha\gamma$ coincidences with $E_\gamma = 404$ keV appear on the $Q[0^+(^{222}\mathrm{Th})\to 2^+(^{218}\mathrm{Ra})]$ line in Fig. 1(a), the state populated by the α decay is likely to subsequently de-excite to the 2^+ state via a 404-keV γ ray. The transition energies from $3^- \to 2^+$ and $2^+ \to 0^+$ in ^{218}Ra have previously been established as 404 keV and 389 keV, respectively, from in-beam studies [10–13]. The 7205-keV α-particle peak is therefore assigned to the decay which directly populates the $J^\pi = 3^-$ state in ^{218}Ra.

C. α decay to the $J^\pi = 1^-$ state

Coincidences between α particles with $E_\alpha = 7143(4)$ keV, and γ rays with energy 853 keV are indicated on Fig. 1(a). Again, the 7143-keV α particles were identified as being from ^{222}Th by their half-life and that of the subsequent ^{218}Ra α decays. Figure 1(c) shows γ rays in coincidence with 7143-keV α-particles; contaminant coincidences are present from escaping α decays of ^{221}Th (576 keV) and ^{219}Ra (592 and 806 keV). The problem of contaminant $\alpha\gamma$ coincidences from ^{213}Rn, which has a similar γ-ray energy, was overcome by requiring a recoil-α-α tag. Figure 1(d) shows the $\alpha\gamma$ spectrum where the ^{213}Rn $\alpha\gamma$ coincidences are removed by the recoil-α-α requirement; six counts in the ^{222}Th $\alpha\gamma$ cluster remain. These coincidences appear on the $Q[0^+(^{222}\mathrm{Th})\rightarrow 0^+(^{218}\mathrm{Ra})]$ line, so it is probable that the state populated by the α decay then de-excites directly to the ground state of ^{218}Ra. This gives a state in ^{218}Ra at 853 keV, which has not previously been observed. No evidence was found that this state at 853 keV decays to the 2^+ state at 389 keV.

The hindrance factor, f, of the α decay populating the 853-keV state is similar to that populating the $J^\pi = 3^-$ state. It is therefore assumed that the two states have a similar underlying structure, as described in Section V A. As no negative-parity state is known at 853 keV it is proposed as a candidate for the $J^\pi = 1^-$ state in ^{218}Ra. This proposed assignment is not in agreement with the previous tentatively assigned $J^\pi = 1^-$ state at 713 keV [10]. Also, population of the 1^- state by α decay would be expected from consideration of the intensity with which the $J^\pi = 3^-$ state is populated. It should be pointed out that no $\alpha\gamma$ coincidences with γ rays of 713 keV were observed in the present data.

V. DISCUSSION

A. α-decay hindrance factors

The hindrance factor of an α decay is the ratio of its experimentally observed partial half-life to the partial half-life calculated using a simple model where the preformed α particle exists in the potential of the daughter nucleus [27]. This eliminates the Q-value dependence of the decay rate and quantifies the relationship between the wavefunctions of the initial state in the mother and the final state in the daughter nuclei; a greater overlap gives a lower hindrance factor. The hindrance factor of a ground-state-to-ground-state α decay for an even-even nucleus is set to unity, meaning that hindrance factors can also be considered as a measure of the similarity of the ground state and excited state of a daughter nucleus populated by an α decay. Figure 3 shows the hindrance factors of α decays to the first $J^\pi = 1^-$ and 3^- states in even-even isotopes of Th, Ra and Rn around the region of expected octupole collectivity [28–34]. Here, the $J^\pi = 1^-$ and 3^- states in ^{218}Rn have been assigned as the 840.2- and 796.9-keV levels respectively, observed following α decay [35]. These levels were not observed using in-beam spectroscopy following a multi-nucleon transfer reaction [36], however, a negative-parity band was established down to $J^\pi = 5^-$. The levels are presently assigned to have $J^\pi = 1^-$ and 3^- from their decay branches to the 0^+, 2^+ and 4^+ members of the ground-state band [35].

If the low-lying negative-parity states were the result of rotation of a reflection-asymmetric nuclear shape then they would be different projections of the reflection-asymmetric ground state. The reduction of the hindrance factors for decreasing N below 140 has therefore previously been interpreted as the onset of intrinsic reflection asymmetry [37]. In this work, a reversal of this trend is observed at $N = 130$ for Ra isotopes, possibly suggesting a departure from static octupole deformations in these nuclei in which the states are no longer described as rotational. This would be consistent with predictions of a near-spherical ground state at $N = 130$ [8] and can be interpreted as a low N boundary to the region of ground-state octupole deformations in the light actinides. An increase in hindrance also occurs at $N = 132$ for the Rn isotopes which has previously been noted in Ref. [38].

B. Relative positions of $J^\pi = 1^-$ and 3^- levels in ^{218}Ra

The possibility of α-particle clustering in the actinides has been proposed in Ref. [39]. In this model, low-lying negative-parity states arise from the mixing of the ground-state quadrupole band and a dipole phonon produced by oscillations between the α-particle cluster and the remaining core. In even-even nuclei this gives positive- and negative-parity states with the $J^\pi = 1^-$ state lying below the 3^-. Energies of the 1^- and 3^- states produced by this α-particle clustering mechanism in Ra and Th were calculated by Daley and Iachello [15] and were shown to agree with the available experimental data for even $^{218-228}$Ra and $^{222-230}$Th. The comparison included the tentative assignment for the $J^\pi = 1^-$ state in ^{218}Ra at 713 keV, 81 keV below the $J^\pi = 3^-$ state [10]; a result contradicted by the present work. The inverted ordering of the $J^\pi = 1^-$ and 3^- levels suggests that neither α-particle clustering or rotation of an asymmetric ground state are responsible for the low-lying negative parity states in ^{218}Ra. In this context, it should also be noted that the anomalously large reduced α-decay width of ^{218}Ra, cited as further evidence for α-particle clustering, has subsequently been contradicted [13, 21, 40, 41].

The evolution of low-lying negative-parity octupole-vibrational states moving from spherical to quadrupole-deformed systems is well understood [42]. In spherical nuclei, negative-parity states can be produced by the $2^+ \otimes 3^-$ multiplet of the coupled quadrupole and octupole vibrational phonons [43, 44]. The $J^\pi = 1^-$

state of the multiplet has $E(1^-) \simeq E(2^+) + E(3^-)$ and therefore appears above the 3^- phonon state. In nuclei with static quadrupole deformation the 3^- octupole-vibrational phonon couples with this deformation [45]. This produces four states with $K^\pi = 0^-, 1^-, 2^-$ and 3^- where K is the projection of the phonon angular momentum onto the nuclear symmetry axis. These states are the band heads of four octupole-vibrational bands, of which those with $K^\pi = 0^-$ and 1^- have a lowest energy state with $J^\pi = 1^-$. Therefore in moving from a spherical to a well-quadrupole-deformed nucleus the relative ordering of the 1^- and 3^- states will reverse.

A study of the systematics of the low-lying negative-parity states observed in both the even-even lanthanides ($Z \simeq 56$, $N \simeq 90$) and light actinides ($Z \simeq 88$, $N \simeq 136$) was carried out by Cottle and Bromley [46]. By plotting $E(3^-) - E(1^-)$ against $E(4^+)/E(2^+)$ for the nuclei in the lanthanide region, it was shown that the behaviour is consistent with that expected for the octupole-vibrational description of the low-lying states, as shown in Fig. 3(a) of Ref. [46]. However, the interpretation of the results for the Rn, Ra and Th nuclei was less conclusive due to the ^{218}Ra data point not matching the expected trend for the octupole-vibrational description. Figure 4 shows the variation of $E(3^-) - E(1^-)$ with $E(4^+)/E(2^+)$ for the nuclei $^{224-232}$Th, $^{218-228}$Ra and $^{218-222}$Rn [28–34] with the present result for ^{218}Ra replacing that tentatively assigned by Gai *et al.* [10]. The new data point is consistent with that expected for an octupole vibrational description of the states in ^{218}Ra, and also across the light actinides.

Predictions by Nazarewicz and Olanders [8] give a picture of octupole vibrations about a spherical nuclear shape for ^{218}Ra and rotation of an asymmetric ground state when increasing the neutron number to $N \simeq 134$. The evolution of the relative positions of the 1^- and 3^- states, as shown in Fig. 4, is not only consistent with the evolution of octupole-vibrational states in an increasingly quadrupole deformed system, but could also be said to be consistent with the predicted onset of rotational states of an octupole-deformed ground state. Evidence for this second scenario is also provided by the evolution of the hindrance factors presented earlier.

VI. SUMMARY

In summary, a state with an excitation energy of 853 keV has been identified in ^{218}Ra and proposed as a candidate for the $J^\pi = 1^-$ state. This observation was made following the identification of α decay of ^{222}Th to both the proposed 1^- state and 3^- state in ^{218}Ra by means of $\alpha\gamma$ coincidence analysis. The hindrance factors of these α decays are larger than those populating analogous states in nuclei with larger N. This then reverses the trend in the Ra isotopes, of decreasing hindrance factor as the neutron number is reduced. These observations are presented as possible evidence for a boundary to the region of static octupole deformations in the radium isotopes. The excitation energy of the 1^- state above that of the 3^- state is presented as an indication that octupole vibrations produce these low-energy levels, as opposed to α-particle clustering or rotations of a reflection-asymmetric ground state. The data for the Th, Ra and Rn isotopes are also consistent with a picture of octupole vibrations of a near-spherical ground state at $N = 130$, moving to rotations of a reflection-asymmetric ground state at $N = 134$.

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TABLE I. α-particle energies, E_α, branching ratios, b_α, and hindrance factors, f, of α decays from the ^{222}Th ground state to the final state J_f^π at an energy E_f in ^{218}Ra.

E_α (keV)	J_f^π	E_f (keV)	b_α (%)	f
7986(3)	0^+	0	98.16(5)	1
7603(3)	2^+	389	1.81(1)	3.38(2)
7205(4)	3^-	793	1.8(3)x10^{-2}	15(3)
7143(4)	(1^-)	853	1.4(4)x10^{-2}	13(4)

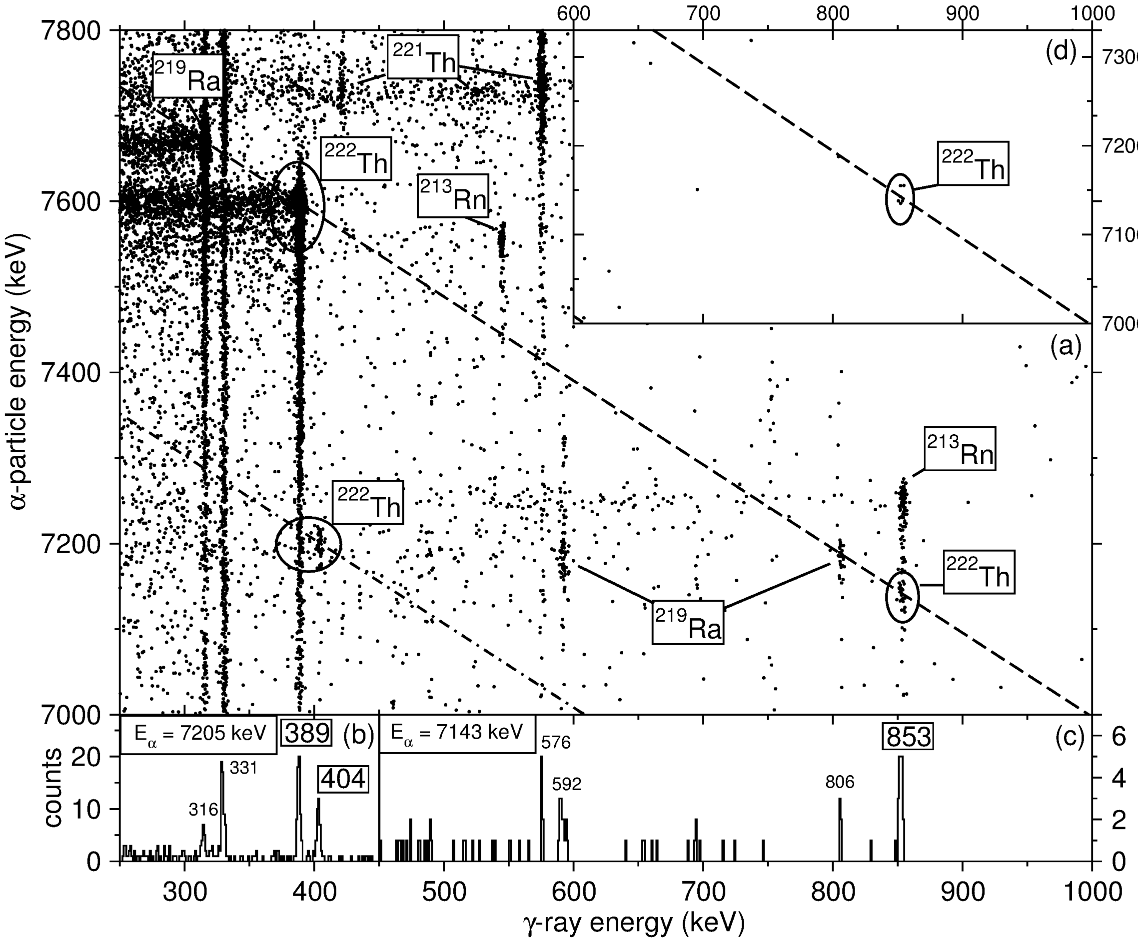

FIG. 1. Energies of coincident α particles and γ rays following the decay of ^{222}Th. Panel (a) shows the coincidences when requiring a recoil-α correlation. The diagonal lines represent a constant energy for the sum of the α-decay Q value, calculated from the α-particle energy, and the γ-ray energy; The energies are those between the ^{222}Th ground state and the ground state (dashed) and 2^+ state (dot dashed) of ^{218}Ra. Panels (b) and (c) show the γ rays in coincidence with α-particle energies of 7205 keV and 7143 keV, respectively. Panel (d) shows the $\alpha\gamma$ coincidences when requiring a recoil-α-α correlation, as discussed in the text.

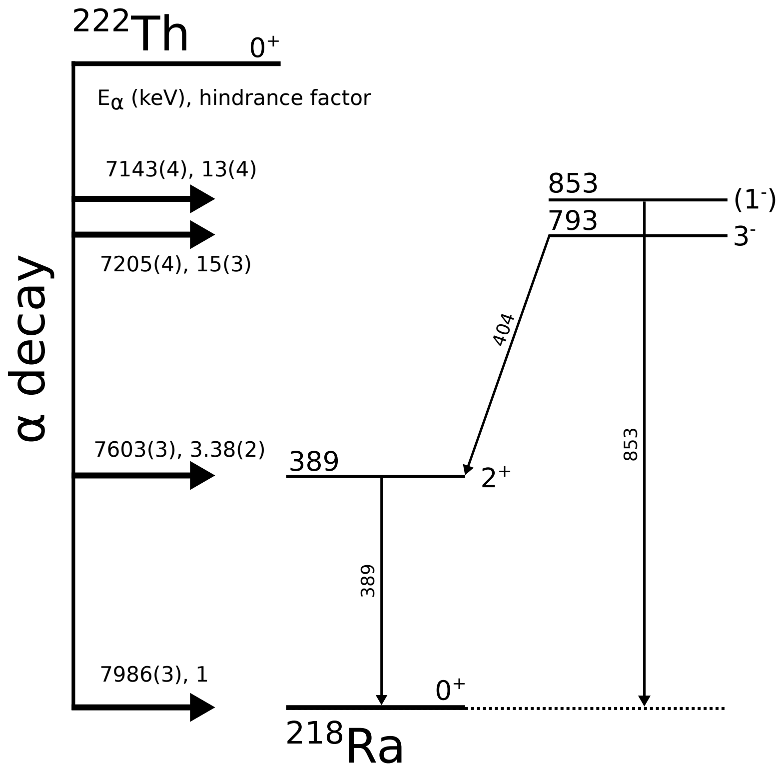

FIG. 2. Level scheme of ^{218}Ra populated following the α decay of ^{222}Th. The α-particle energies, E_α, and hindrance factors are indicated. The 1^- level proposed in this work is shown at 853 keV.

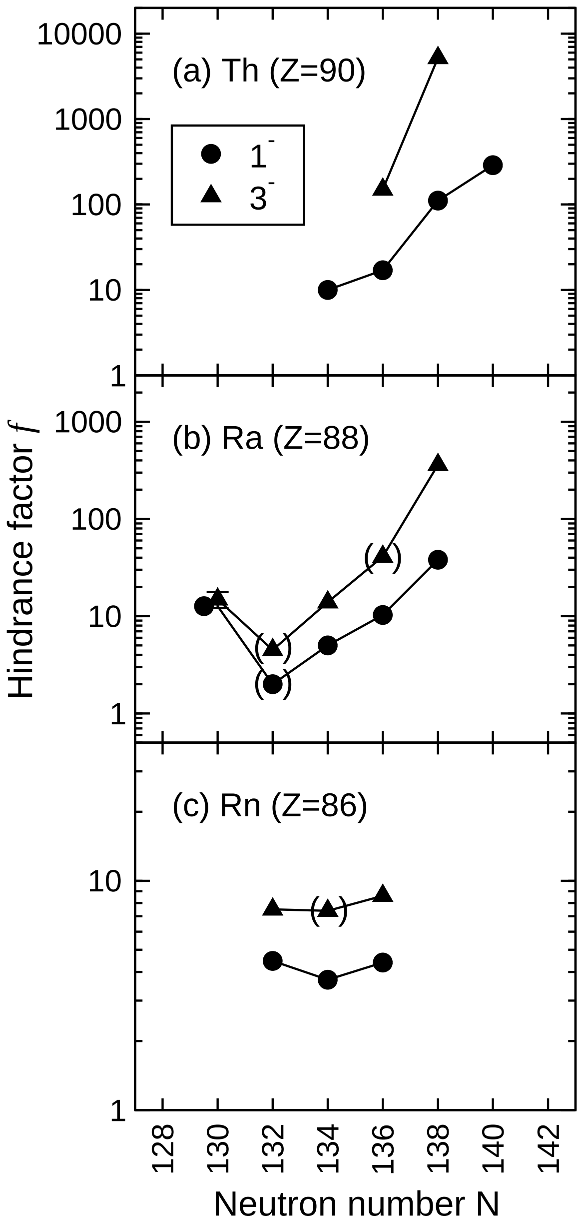

FIG. 3. Hindrance factors of the α decays populating the 1^- and 3^- states in (a) Th, (b) Ra and (c) Rn isotopes as a function of neutron number N [28–34]. The 1^- data point for Ra at $N = 130$ has been shifted to the left for clarity; data points in brackets are tentative.

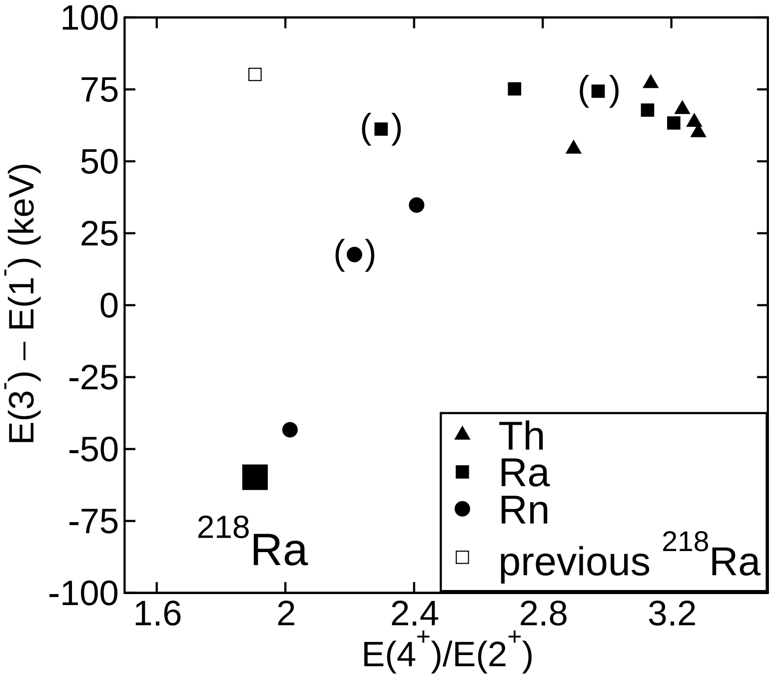

FIG. 4. Relative positions of the 1^- and 3^- states in even-even $^{224-232}$Th, $^{218-228}$Ra and $^{218-222}$Rn nuclei [28–34] compared with the ratio of their first 4^+ and 2^+ state energies, $E(4^+)/E(2^+)$. The new value for ^{218}Ra is enlarged and the previously assigned value is shown as an open symbol; data points in brackets are tentative.

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